

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE



File Application of

MOCK ET AL

Serial No. 09/361,626

Group Art Unit: 1724

Filed: July 27, 1999

Examiner: Cintins

For: PROCESS FOR REMOVING
ALUMINUM SPECIES FROM
ALKALI METAL HALIDE BRINE
SOLUTIONS

DECLARATION OF ZBIGNIEW TWARDOWSKI

Hon. Commissioner of
Patents and Trademarks
Washington, D.C. 20231

Sir:

I, Zbigniew Twardowski, declare as follows:

1. My higher education includes:

Itemized in the attached resume

2. My employment history comprises:

Itemized in the attached resume and List of Publications/Patents

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3. Attached hereto is my curriculum vitae showing my educational background and employment history as well as listing papers I have authored, patents granted and honors received.

4. I have studied the above-identified U.S. patent application Serial No. 09/361,626 and the history of its examination in the U.S. Patent and Trademark Office. This has included a review of the Examiner's Office Actions, the applicants' responses and the cited prior art, particularly, Nagy U.S. Patent 4,073,706 (assignee: Diamond Shamrock).

5. The invention disclosed and claimed in Serial No. 09/361,626 is concerned with the reduction of soluble aluminum species in an alkali metal brine to provide a brine feedstock suitable for use in a chlor-alkali cell process. It is critically important to remove aluminum species from such feedstock for a variety of reasons including, in particular, environmental considerations. However, despite considerable research effort directed towards removal of aluminum from brine, the presence of small amounts of aluminum in brine to be used electrolytically has remained a serious problem. The method described and claimed in Serial No. 09/361,626 represents an important advance towards solving the problem.

6. According to the invention represented by Serial No. 09/361,626, the brine is treated with a magnesium salt in amount sufficient to provide a Mg/Al molar ratio of 5-20 to 1 and a Mg concentration of from 0.5 to less than 5 ppm. Sufficient alkali metal hydroxide is included to provide an excess alkalinity concentration of between 0.1 and 0.3 grams per liter alkali metal hydroxide. This precipitates a magnesium aluminum hydroxide complex, the latter being removed to provide the desired brine feedstock.



7. Essential to the success of the invention is the observance of the indicated Mg/Al molar ratio, and Mg and alkali metal hydroxide concentrations. The applicants have found that the observance of these conditions results in a rapid and effective removal of soluble aluminum species to provide a highly suitable brine feedstock for use in a chlor-alkali cell process.

8. I am aware that the invention of Serial No. 09/361,626 is defined as follows in claim 1 of the application:

1. A method for the reduction of soluble aluminum species in an evaporated salt alkali metal halide brine containing up to 500 ppb aluminum species to provide a brine feedstock suitable for use in a chlor-alkali membrane cell process, said method comprising:

(a) treating said brine with a magnesium salt in an amount to provide a Mg to Al molar ratio selected from 5-20 to 1 and at a Mg concentration of at least 0.5 and less than 5 ppm, and sufficient alkali metal hydroxide to provide an excess alkalinity concentration of between 0.1-0.3 g/L alkali metal hydroxide to effect precipitation of a magnesium aluminum hydroxide complex; and

(b) removing said complex to provide said brine feedstock.

9. I understand that the U.S. Examiner has rejected claim 1 and the other claims pending in Serial No. 09/361,626 as unpatentable over Nagy, U.S. Patent 5,073,706 (Diamond Shamrock). I am fully familiar with the Nagy patent and I note that the patent discloses a process for reducing the amount of trace metals, e.g. aluminum, in aqueous alkali halide brine. This is done by the addition of magnesium ions to the brine and subsequent precipitation of magnesium hydroxide to remove metal contaminants and provide a brine suitable for use in the electrolytic production of chlorine and alkali metal hydroxide. According to the patent, a soluble magnesium compound, e.g. magnesium chloride, is added to the brine solution to provide an Mg concentration of greater than 5 ppm. The pH of the brine is also adjusted to about 8 to 12 by adding NaOH thereto.

10. I note that the Examiner in Office Action of December 7, 2000 (Paper No. 10) has acknowledged that the Nagy patent does not disclose the applicants' recited magnesium to aluminum molar ratio, or the magnesium concentration in the brine (i.e. less than 5 ppm). Nevertheless, the Examiner concluded that the

"exact magnesium to aluminum molar ratio in the reference process is not seen to materially affect the overall results of this process, or to produce any new and unexpected result; and is therefore deemed to be an obvious matter of choice, which is insufficient to patentably distinguish to the claims. Furthermore, it would have been obvious to one of ordinary skill in the art at the time the invention was made to employ a brine having a magnesium concentration slightly lower than that disclosed (e.g. 4.99 ppm) in the reference, since such a slightly lower magnesium concentration is also not seen to materially affect the overall results of the reference process, or to produce any new and unexpected result; and is therefore also deemed to be an obvious matter of choice, which is insufficient to patentably distinguish the claims."

The Examiner repeated this position in the subsequent Office Action of June 5, 2001 (Paper No. 12, pages 2-3).

11. The applicants in Serial No. 09/361,626, however, have discovered that there is a critical relationship in the Mg to Al molar ratio, Mg concentration (less than 5 ppm), and use of alkali metal hydroxide to provide the excess alkalinity recited in claim 1 (0.1 to 0.3 g/L) if the most effective aluminum removal is to be obtained. Stated otherwise, the applicants have found that they obtain unexpected results, in terms of aluminum removal efficiency, as high as 99% removal over short treatment times, by observing the requirements of their claim 1 regarding molar ratio of Mg to Al, Mg concentration and alkali metal hydroxide excess.

The unexpected results obtainable with the applicants' invention are shown in attached Exhibit A. This Exhibit sets forth experiments and the results thereof, including charts, wherein conditions representative of the applicants' method and Nagy's method have



been compared. Exhibit A includes the results of three experiments, each of which includes nine separate runs or tests and a further experiment comprising eight different runs considered reasonably representative of Nagy. The various runs constituting Experiment Nos. 1-3 and the Experiment representative of Nagy were carried out under the same conditions except for the variations in the columns representing Al content in the starting brine, Mg added, Mg/Al molar ratio and NaOH. Removal efficiency was measured at the intervals (residence times) indicated.

The various tests were carried out by adding Mg and NaOH to brine containing Al at 50°C. This resulted in the precipitation of a complex of Mg, Al and OH which was removed, after which the resulting brines were checked for efficiency of Al removal. In Experiment Nos. 1-3, efficiency was measured immediately after addition of the Mg and NaOH and removal of the precipitate (i.e. zero residence time) and after 15 and 30 minutes residence times. The residence times for the Nagy experiment were 20 minutes, 40 minutes and 180 minutes at 66°C. The results of the experiments are tabulated in Exhibit A and graphically illustrated by the charts attached thereto. These results show how the efficiency of Al removal varies with Mg content, Mg/Al ratio and alkali concentration (NaOH).

The "Conclusion" section (pages 3-4 of Exhibit A) brings out the importance of observing the specific combination of conditions called for in the applicants' claims. This combination of conditions is not disclosed or suggested by Nagy (Diamond Shamrock) and the results thereof could not have been predicted from Nagy.

12. While the results set forth in Exhibit A speak for themselves in showing the unpredictable results obtained by the applicants' invention, the following specific comments on the results given in Exhibit A may be useful:



(a) Of the Experiment No. 1 runs or tests, only the first two meet the requirements of the applicants' invention regarding Mg added, Mg/Al molar ratio and NaOH added. The remaining runs of Experiment No. 1 use either more NaOH (the third run) and/or a lower Mg/Al molar ratio (all the other runs).

The test results show that the first two runs, i.e. those consistent with the applicants' invention, give the highest aluminum removal efficiency after 15 and 30 minutes. Comparison of the first and fourth runs of Experiment No. 1, which differed only in having a Mg/Al molar ratio of 5.6 (according to the invention) or 2.8, respectively, shows a significant difference in % removal efficiency. The same is true with respect to the 2nd and 5th runs which again differ only in the Mg/Al molar ratio.

(b) The tabulated results for Experiment Nos. 2 and 3 similarly show better removal efficiency using the applicants' conditions (runs 1, 2, 4, 5 of Experiment No. 2 and runs 4, 5 of Experiment No. 3) compared with other conditions where the Mg/Al ratios and/or NaOH additions fall outside the applicants' invention. While it is true that, for example, the removal efficiency obtained in the 4th and 5th runs of Experiment No. 2, which are representative of the applicants' invention, are lower than those obtained in the third run, which is outside the invention because of the higher NaOH content, I do not view this as a proper comparison because the third run includes twice the Mg/Al ratio. The proper comparison is with the sixth run and it is to be noted that the 4th and 5th runs give better efficiencies than the sixth run.

(c) The results of Experiment No. 3 are particularly striking as these show that runs representative of the invention (the 4th and 5th runs where the Mg/Al molar ratio is 11.1 and the NaOH content is 0.1 or 0.2) gave higher removal efficiencies than comparable runs (first and second) using double the Mg/Al ratio (22.2). While the removal efficiency for



the first, second and third runs of Experiment No. 3 is good, the possibility of obtaining equivalent or, in this case, better removal efficiency at the lower Mg/Al ratio (11.1) consistent with the applicants' invention minimizes the possibility of introducing a further unwanted solids to the brine feedstock.

13. With respect to the Experiment representative of Nagy in Exhibit A, it is noted that the use of an Mg/Al molar ratio of 2.8 with initial Al concentration at 1.0 ppm and Mg content of 2.5 ppm resulted in no removal of Al after 20 minutes. This is consistent with the Nagy disclosure which teaches that a magnesium level below 5 ppm is ineffective for removing undesirable metallic impurities. However, Experiment Nos. 1-3 show that effective aluminum removal can be realized when the Mg content is below 5 ppm if the Mg/Al molar ratio and NaOH content are appropriately chosen. In fact, Experiment No. 1 even shows some Al removal with a molar ratio of 2.8 under certain conditions although the results are not nearly as good as those obtained with the applicants' invention. The experiment representative of the Nagy patent shows that the method requires the use of larger concentrations of Mg with consequently higher Mg/Al molar ratios than contemplated by the applicants in Serial No. 09/361,626 with the prospective problem, at the higher Mg concentrations, of introducing undesired solids into the brine.

14. The following additional test results further illustrate the advantages of the invention of Serial No. 09/361,626 and the criticality of the conditions used therein: Two experiments were carried out on brine of ion-exchange quality to determine aluminum removal efficiency under various conditions involving the addition of magnesium (as magnesium chloride) and NaOH to the brine. The temperature of the brine was maintained at 50-60°C and the residence time was 5-15 minutes. Experiment #1 included three separate tests on brine containing 0.5 ppm of aluminum. The amount of Mg added to the three brine

samples was 3 ppm, 4 ppm and 5 ppm, respectively, giving a Mg/Al molar ratio in these tests of 6.7, 8.9 and 11.1, respectively. NaOH was also added to provide an excess thereof of between about 0.1-0.2 g/l. After 15 minutes, the magnesium aluminum complex which separated out was removed and the residual amount of aluminum in the resulting brine was determined in parts per million (ppm). The efficiency of aluminum removal for each test is shown in Table 1 below.

TABLE 1

Initial Aluminum Concentration (ppm)	Magnesium Concentration Added (ppm)	Mg/Al Molar Ratio	Residual Aluminum (ppm)	Al Removal Efficiency (%)
0.5	3	6.7	0.010	98
0.5	4	8.9	0.001	99
0.5	5	11.1	0.001	99

It will be noted from Table 1 that by maintaining a Mg/Al molar ratio as shown, Al removal efficiencies of 98% or higher were realized.

The conditions used in the three tests comprising Experiment #1 were all within the applications' invention.

Experiment #2 was carried out as in Experiment #1 except that Experiment #2 involved five separate tests on brine containing 1 ppm of aluminum to which there was added 1, 2, 3, 4 and 5 ppm, respectively, of magnesium to provide Mg/Al molar ratios of 1.1, 2.2, 3.3, 4.4 and 5.5, respectively. Because of the different Mg/Al molar ratios, only the fifth test was within the applicants' invention, the Mg/Al molar ratios for the other tests in Experiment #2 being below the ratio the applicants require.

After the addition of Mg and NaOH to each brine specimen, the five brines of Experiment #2 were allowed to stand as in Experiment #1 after which the precipitated Mg/Al complex was removed and the brine checked for residual aluminum. The results are shown in Table 2 below:

TABLE 2

Initial Aluminum Concentration (ppm)	Magnesium Concentration Added (ppm)	Mg/Al MolarRatio	Residual Aluminum (ppm)	Al Removal Efficiency (%)
1	1	1.1	0.840	16
1	2	2.2	0.650	35
1	3	3.3	0.430	57
1	4	4.4	0.210	79
1	5	5.5	0.010	99

15. As will be evident, only the last test shown in Table 2 is according to the applicants' invention. This test gave an Al removal efficiency of 99% and substantially higher than the removal efficiencies obtained with the other tests, all of which were outside the applicants' invention because of the lower Mg/Al mol ratios used therein.

16. The results referred to above and illustrated in the attached Exhibit A establish, in my opinion, criticality in Mg and NaOH concentrations, and Mg/Al molar ratios as defined in claim 1 of Serial No. 09/361,626 when one is concerned with removing Al from brine to prepare a brine feedstock for chlor-alkali electrolysis cells. This criticality is not disclosed in, or evident from, the Nagy U.S. patent and, in my opinion, the applicants' finding as described and claimed in Serial No. 09/361,626 is not in any sense obvious.



17. I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both under § 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

E. Tranter

Date: April 24, 2002

Exhibit "A" to Declaration
of Zbigniew Twardowski

ZBIGNIEW TWARDOWSKI

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GENERAL SUMMARY

P.Eng., Ph.D. Chemistry, M.Eng., Chemical Engineering. Over 20 years of experience in membrane separation technologies and development of novel, primarily electrochemical processes. Proven R&D leadership and strategy skills. Awarded 16 patents (incl. 3 commercialized technologies).

CAREER OBJECTIVE: Management of technology development & transfer leading to new business.

PROFESSIONAL EXPERIENCE

1992 – **R&D Manager, Kvaerner Chemetics, Vancouver, B.C., Canada**

Responsibilities: Directs R&D Department oriented towards development of proprietary process technologies, primarily in membrane separation and advanced electrochemistry areas. Member of the Company's Management Committee. Provides leadership in identification and screening of new technical/commercial opportunities. Manages Company's patent portfolio. Initiates and administers numerous external R&D programs with outside/offshore companies, research institutes and universities. Successfully secured government grants for research projects. Invented and developed the commercialized SRS technology, which won the 1999 Filtration & Separation Product Achievement Award (Membrane Application Category) as well as the 2001 Environmental Award of The Assoc. of Professional Engineers and Geoscientists of B.C.. To-date some 13 SRS plants were built worldwide.

1990 – 1992 **Manager, R&D New Technologies, Albright & Wilson Americas, Toronto,**

Responsibilities: Supervised R&D Group of 6 professionals mandated to provide "technology push" for A&WA Pulp Chemicals Division and, especially, for the ERCO Chemical Systems Group. Developed and piloted major patented processes: Saltcake Electrolysis (R9 Process), and Electrosynthesis of Chloric Acid. Successfully used gas diffusion electrodes in electrosynthesis.

1984 – 1990 **Research Manager/Assistant R&D Manager, A&WA, Toronto**

Responsibilities: Supervised a group of 10 –14 professionals and technologists engaged in development of electrochemical processes, primarily for the new bleaching agents and of novel membrane separation processes. Directed R&D work in support of the existing A&WA business, i.e. Sodium Chlorate/Chlorite manufacture and Chlorine Dioxide Generation. Developed internal expertise in 3-D electrolyzer design, which resulted in successfully piloted processes for Arsenic Removal and Electrosynthesis of Sodium Chlorite. Actively involved in assessments of new commercial opportunities for the Company such as manufacturing of Persulfates, Hydrogen Peroxide, Sodium Hydrosulfite, Sodium Borohydride, Anthraquinone and fabrication of Bipolar Membranes.

1981 – 1984 **Research Chemist/Senior Research Chemist**, A&WA, Toronto

Responsibilities: Functioned as Project Leader, novel electrochemical processes, new cell designs, membrane separations (Dialysis, Electrodialysis, Bipolar Membranes, Gas Transfer, Liquid Membranes). Developed practical methods for effluent treatment in phosphoric acid plants. Consulted in the areas of sodium chlorate and chlorine dioxide technologies. Conducted pilot plant trials to generate data for process design.

1980 – 1981 **Research Associate**, Department of Chemistry, The University of Calgary, Calgary, Alberta, Canada.

Co-researcher (to Prof. H. L. Yeager) in investigation of morphology and transport properties of the Nafion brand ion-exchange membranes in hot concentrated electrolyte solutions, typical of Chloralkali and Water Electrolysis processes.

1973 – 1974 **Research Officer**, Central Fishery Laboratory, Gdynia, Poland

Responsible for development projects in corrosion prevention of tinplated and aluminum containers and application of modern chemical control instrumentation in fish processing. In 1976 dispatched by the employer to USA for advanced (Ph.D.) studies.

EDUCATION

1976 – 1980 **Ph. D., Analytical Chemistry**, Southern Illinois University at Carbondale, Carbondale, Illinois, USA

Thesis Research: Application of ion exchange membranes to chemical separations and metal speciation in aqueous systems.

Techniques Used: Donnan Dialysis, Modern Voltammetric Methods, AAS, FAAS.

Awards: Dissertation Award from SIU-C, Summer 1980.

1968 – 1973 **M.Sc. (Mgr. Ing.), Chemical Technology of Inorganic Compounds**, Technical University of Gdansk, Gdansk, Poland

Specialization: Protection Against Corrosion

Intensive Courses Taken: Effective Communication, R&D Management, Hazards and Operability, Industrial Electrochemistry, Membrane Summer School, Economic Project Analysis, Total Quality Management (several)

MEMBERSHIP IN PROFESSIONAL ORGANIZATIONS

- Professional Engineers - Ontario
- The Electrochemical Society, Canadian Section, **Chairperson in 1991/1992**
- North American Membrane Society
- European Membrane Society

Zbigniew Twardowski

Page Three

PERSONAL

Married, Two Children
Excellent Health, Active/enjoy Travel, Camping, Tennis, Bridge
Fluent in English, Polish and Russian

REFERENCES

Available upon request

Zbigniew Twardowski

LIST OF PUBLICATIONS

1. J. A. Cox and Z. Twardowski, "Donnan Dialysis Matrix Normalization for the Voltammetric Determination of Metal Ions", **Analytica Chimica Acta**, **119**, 39-45 (1980).
2. J. A. Cox and Z. Twardowski, "Tubular Flow Donnan Dialysis", **Analytical Chemistry**, **52**, 1503-1505 (1980).
3. J. A. Cox and Z. Twardowski, "Electric Field Enhancement of Donnan Dialysis", **Analytical Letters**, **13(A14)**, 1283-1291 (1980).
4. H. L. Yeager, B. O'Dell and Z. Twardowski, "Transport Properties of Nafion Membranes in Concentrated Solution Environments", **J. Electrochem. Soc.**, **129**, 85-89 (1982).
5. H. L. Yeager, Z. Twardowski and L. Clarke, "A Comparison of Perfluorinated Carboxylate and Sulfonate Ion Exchange Polymers. I. Diffusion and Water Sorption", **J. Electrochem. Soc.**, **129**, 324-327 (1982).
6. Z. Twardowski, H.L. Yeager and B. O'Dell, "A Comparison of Perfluorinated Carboxylate and Sulfonate Ion Exchange Polymers. II. Sorption and Transport Properties in Concentrated Solution Environments", **J. Electrochem. Soc.**, **129**, 328-332 (1982).
7. J. A. Cox, T. Gray. K. S. Yoon and Z. Twardowski, "Selection of the Receiver Electrolyte for the Donnan Dialysis Enrichment of Cations", **Analyst**, **109**, 1603-1605 (1984).
8. V. I. Volkov, Yu M. Popkov, S. F. Timashev, D. G. Bessarabov, R. D. Sanderson and Z. Twardowski, "Self-diffusion of water and fluorine ions in anion-exchange polymeric materials, as determined by pulse-field nuclear magnetic resonance spectroscopy", **J. Membrane Science**, **180**, 1-13 (2000).

and some 50 proprietary research reports

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1. J. A. Cox, Z. Twardowski and K. H. Cheng, "Donnan Dialysis Matrix Normalization for Stripping Voltammetry Analysis", **Joint Central-Great Lakes Regional ACS Mtg., Indianapolis, IN, 1978**.
2. J. A. Cox, Z. Twardowski and G. R. Litwinski, "Donnan Dialysis Sampler For Continuous On-Stream Metal Determinations", **Paper #551, Pittsburgh Conference, Atlantic City, 1981**.
3. H. L. Yeager and Z. Twardowski, Measurement of Ionic and Water Transport Numbers in Membrane Chloralkali Cell", **Spring Meeting of the Electrochemical Society, Minneapolis, 1981**.
4. G. J. Ranger and Z. Twardowski, "Combined Electrochemical Process for Production of Chlorine Dioxide and Caustic", **3rd Intl. Forum on Electrolysis In Chemical Industry, Ft. Lauderdale, FL, 1989**.
5. G. Cowley, M. Lipsztajn and Z. Twardowski, "Electrolysis of Chloric Acid at High Current Efficiencies", **5th Intl. Forum on Electrolysis In Chemical Industry, Ft. Lauderdale, FL, 1991**.

6. Z. Twardowski et al, "An Improved Anode Coating For Electrosynthesis of Sodium Chlorate", **7th Intl. Forum on Electrolysis in Chemical Industry, Lake Buena Vista, FL, 1993.**
7. A. Bacon, Z. Twardowski and N. Tam, "Improved Anode Coatings for Elevated Temperature Operation of Sodium Chlorate Cells", **Intl. Chlorine Symposium, London, UK, 1994.**
8. K. Maycock, Z. Twardowski and J. Ulan, "A New Method to Remove Sulphate From Brine", **Intl. Chlorine Symposium, London, UK, 1997.**
9. Z. Twardowski, C. Kotzo, J. Ulan and K. Maycock, "A New Method to Remove Sulphate from Concentrated Brine", **11th Intl. Forum on Electrolysis in Chemical Industry, Clearwater Beach, FL, 1997.**
10. V. D. Jovic, B. M. Jovic, G. R. Stafford, N. V. Krstajic and Z. Twardowski, "Composition and Morphology Changes and their Influence on Hydrogen Evolution on Ni-Mo and Fe-Mo Alloys Electrodeposited by DC and Pulsating Regimes", **AESF SURF/FIN, Chicago, IL, June 2002.**

PATENTS

1. Z. Twardowski, "Method of Regulation of Concentration of Acid or Base", **Polish Patent 103,938, 1976.**
2. Z. Twardowski, "Apparatus for Regulation of Concentration of Acid or Base", **Polish Patent 112,427, 1978.**
3. Z. Twardowski, "Acid Recovery in Chlorine Dioxide Concentration", **U.S. Patent 4,678,655, 1987.**
4. Z. Twardowski and J. D. McGilvery, "Membrane Perstraction Process for Chlorine Dioxide", **U.S. Patent 4,683,039, 1987.**
5. Z. Twardowski, "Removal of Arsenic from Acids", **U.S. Patent 4,692,228, 1987.**
6. Z. Twardowski, "Combined Process for Production of Chlorine Dioxide and Sodium Hydroxide", **U.S. Patent 4,806,215, 1989.**
7. M. Lipsztajn, J. D. McGilvery and Z. Twardowski, "Production of Chlorine Dioxide in An Electrolytic Cell", **U.S. Patent 4,915,927, 1990.**
8. M. Lipsztajn, Z. Twardowski and G. Cowley, "Electrochemical Production of Chloric Acid and Caustic", **U.S. Patent 4,915,927, 1990.**
9. G. Cowley, M. Lipsztajn and Z. Twardowski, "Electrochemical Processing of Aqueous Solutions", **U.S. Patent 5,122,240, 1992.**
10. Z. Twardowski, M. Lipsztajn and M. C. Fredette, "Chlorine Dioxide Generation from Chloric Acid", **U.S. Patent 5,174,868, 1992.**
11. G. Cowley, M. Lipsztajn, Z. Twardowski, R. Swindells and E. J. Bechberger, "Electrochemical Processing of Aqueous Solutions", **U.S. Patent 5,198,080, 1993.**
12. Z. Twardowski, "Method of Membrane Cell Washing", **U.S. Patent 5,277,768, 1994.**
13. M. Lipsztajn, M. C. Fredette, Z. Twardowski and E. J. Bechberger, "Chlorine Dioxide Generation From Chloric Acid", **U.S. Patent 5,284,553, 1994.**

14. Z. Twardowski, "Nanofiltration of Concentrated Salt Solutions", **U.S. Patent 5,587,083, 1996.**
15. Z. Twardowski and J. G. Ulan, "Nanofiltration of Concentrated Salt Solutions", **U.S. Patent 5,858,240, 1999.**
16. N. Tam, Z. Twardowski and M. Geusic, "Metal Electrode for Electrochemical Processes", **Canadian Patent 2,166,494, 2001.**

BRITISH PRELIMINARY PATENT APPLICATIONS

1. M. Lipsztajn and Z. Twardowski, "Electrogeneration of Ozone Using Solid Polymer Electrolyte Based Fluoroanion Containing Membrane", **U.K. 8700412 (January 8, 1987).**
2. Z. Twardowski and H. Twardowska, "Method of Production of Hypophosphorous Acid", **U.K. 8706805 (March 23, 1987).**

PENDING PATENT APPLICATIONS

1. Z. Twardowski, F. Mok and T. Drackett, "Gas Diffusion Electrode for Electrosynthesis of Hydrogen Peroxide".

Optimization of Aluminum Removal by Magnesium Chloride Addition

The following results have been tabulated to illustrate the effects of Aluminum removal using Magnesium Chloride at various operating conditions.

Temperature: 50°C
 Magnesium Chloride & NaOH: Reagent Grade Quality
 Brine: Ion-Exchanged Quality

Experiment #1				Residence Time (Minutes)					
Al (ppm)	Mg (ppm)	Mg/Al Molar Ratio	NaOH (g/L)	0		15		30	
				Residual Al (ppm)	Removal Efficiency (%)	Residual Al (ppm)	Removal Efficiency (%)	Residual Al (ppm)	Removal Efficiency (%)
0.1	0.5	5.6	0.1	0.064	36	0.034	66	0.028	72
0.1	0.5	5.6	0.2	0.046	54	0.044	56	0.039	61
0.1	0.5	5.6	0.5	0.055	45	0.055	45	0.049	51
0.2	0.5	2.8	0.1	0.130	41	0.119	46	0.115	48
0.2	0.5	2.8	0.2	0.139	37	0.130	41	0.129	41
0.2	0.5	2.8	0.5	0.173	21	0.139	37	0.139	37
0.5	0.5	1.1	0.1	0.390	22	0.390	22	0.380	24
0.5	0.5	1.1	0.2	0.430	14	0.430	14	0.440	12
0.5	0.5	1.1	0.5	0.440	12	0.410	18	0.420	16

Experiment #2

Al (ppm)	Mg (ppm)	Mg/Al Molar Ratio	NaOH (g/L)	Residence Time (Minutes)			
				0	15	30	
				Residual Al (ppm)	Removal Efficiency (%)	Residual Al (ppm)	Removal Efficiency (%)
0.1	1.0	11.1	0.1	0.009	91	0.009	91
0.1	1.0	11.1	0.2	0.013	87	0.010	90
0.1	1.0	11.1	0.5	0.028	72	0.018	82
0.2	1.0	5.6	0.1	0.065	68	0.070	65
0.2	1.0	5.6	0.2	0.103	49	0.094	53
0.2	1.0	5.6	0.5	0.109	46	0.097	52
0.5	1.0	2.2	0.1	0.268	46	0.251	50
0.5	1.0	2.2	0.2	0.332	34	0.309	38
0.5	1.0	2.2	0.5	0.362	28	0.363	27

Experiment #3

Al (ppm)	Mg (ppm)	Mg/Al Molar Ratio	NaOH (g/L)	Residence Time (Minutes)			
				0	15	30	
				Residual Al (ppm)	Removal Efficiency (%)	Residual Al (ppm)	Removal Efficiency (%)
0.1	2.0	22.2	0.1	0.012	88	0.015	85
0.1	2.0	22.2	0.2	0.011	89	0.012	88
0.1	2.0	22.2	0.5	0.010	90	0.009	91
0.2	2.0	11.1	0.1	0.020	90	0.014	93
0.2	2.0	11.1	0.2	0.033	84	0.017	92
0.2	2.0	11.1	0.5	0.055	73	0.023	89
0.5	2.0	4.4	0.1	0.175	65	0.162	68
0.5	2.0	4.4	0.2	0.194	61	0.209	58
0.5	2.0	4.4	0.5	0.268	46	0.252	50

Diamond Shamrock Corporation (Zoltan Nagy) U.S. Patent No. 4,073,706

Temperature : 66°C

Al (ppm)	Mg (ppm)	Mg/Al Molar Ratio	NaOH (g/L)	20 Minutes of Residence Time	
				Residual Al (ppm)	Removal Efficiency (%)
1.0	2.5	2.8	0.1	1.0	0
1.0	15	16.7	0.5	0.14	86
1.0	62	68.8	0.5	0.26	74
1.0	122	135.5	0.5	0.16	84

Al (ppm)	Mg (ppm)	Mg/Al Molar Ratio	NaOH (g/L)	60 Minutes of Residence Time	
				Residual Al (ppm)	Removal Efficiency (%)
1.0	15	16.7	0.5	0.3	70

Al (ppm)	Mg (ppm)	Mg/Al Molar Ratio	NaOH (g/L)	180 Minutes of Residence Time	
				Residual Al (ppm)	Removal Efficiency (%)
1.0	68	75.5	0.9	0.08	92
1.0	146	162.1	0.9	0.06	94
1.0	122	135.5	0.5	0.04	96

Conclusion:

Aluminum removal efficiency of 90% has been successfully demonstrated when only 1 ppm and 2 ppm of Mg in the form of $MgCl_2$ was added to a brine solution containing 0.1 and 0.2 ppm Al with 0.1 to 0.2 g/L excess NaOH. This translates to a Mg to Al molar ratio of 1.1 to 1. Refer to Experiment #2 & #3 data tables. Furthermore, there is also a delicate balance existing between the excess alkalinity level and the Magnesium concentration in the brine solution.

Once all the Magnesium ions have been consumed to forming the gelatinous hydroxide precipitate (amphoteric nature), the excess hydroxide anions tend to re-solubilize the complex precipitate thus releasing the Aluminum impurities back into solution. This effect is clearly indicated by the lower Aluminum removal efficiency at the higher excess alkalinity concentrations. Refer to Experiment #1, #2 & #3 data tables.

The reaction kinetics of this complex precipitation mechanism is instantaneous where upon only traces of Magnesium, 0.5 to 2.0 ppm, in the presence of 0.1 to 0.2 g/L NaOH, the removal of Aluminum is almost immediate and increase in residence time has not significantly improved on the overall removal efficiency.

The Diamond Shamrock Patent clearly cites that Magnesium level of less than 5 ppm concentration have been found to be ineffective in removing many of the undesirable metallic impurities including Aluminum, as indicated in its example where zero Aluminum removal efficiency was attained in a brine solution containing 1 ppm Al with using 2.5 ppm Mg and 0.1 g/L NaOH excess alkalinity after 20 minutes of reaction time. The working ratio of Mg to Al for this particular example translates to 2.8 to 1. When compared to a similar working Mg/Al ratio in our claims but using only 0.5 to 1.0 ppm Mg, a 50% Aluminum removal efficiency was achieved. Refer to Experiment #1 & #2 data tables and Chart #1 & #2.

This Aluminum removal process using low levels of Magnesium concentration has been successfully operated at two separate chloralkali plants in the United States. The Chloromone Co. in Delaware City was the first plant to fully utilize the benefits of this technology. It was commissioned back in January 1999 with maximum capacity of 50 stpd of chlorine achieved after one month of operation. The second plant which has adopted the identical treatment process was Odyssey Manufacturing Co. located in Tampa, Florida. The chloralkali facility was commissioned back in March 2000 with completion of performance test achieved after only two weeks of operation.

Chart 1 - Comparison of Aluminum Removal Efficiency (%) at Various Mg/Al Molar Ratio with
 Low Magnesium Concentration
 Temperature at 50 to 66°C; Excess Alkalinity Level at 0.5 g/L NaOH

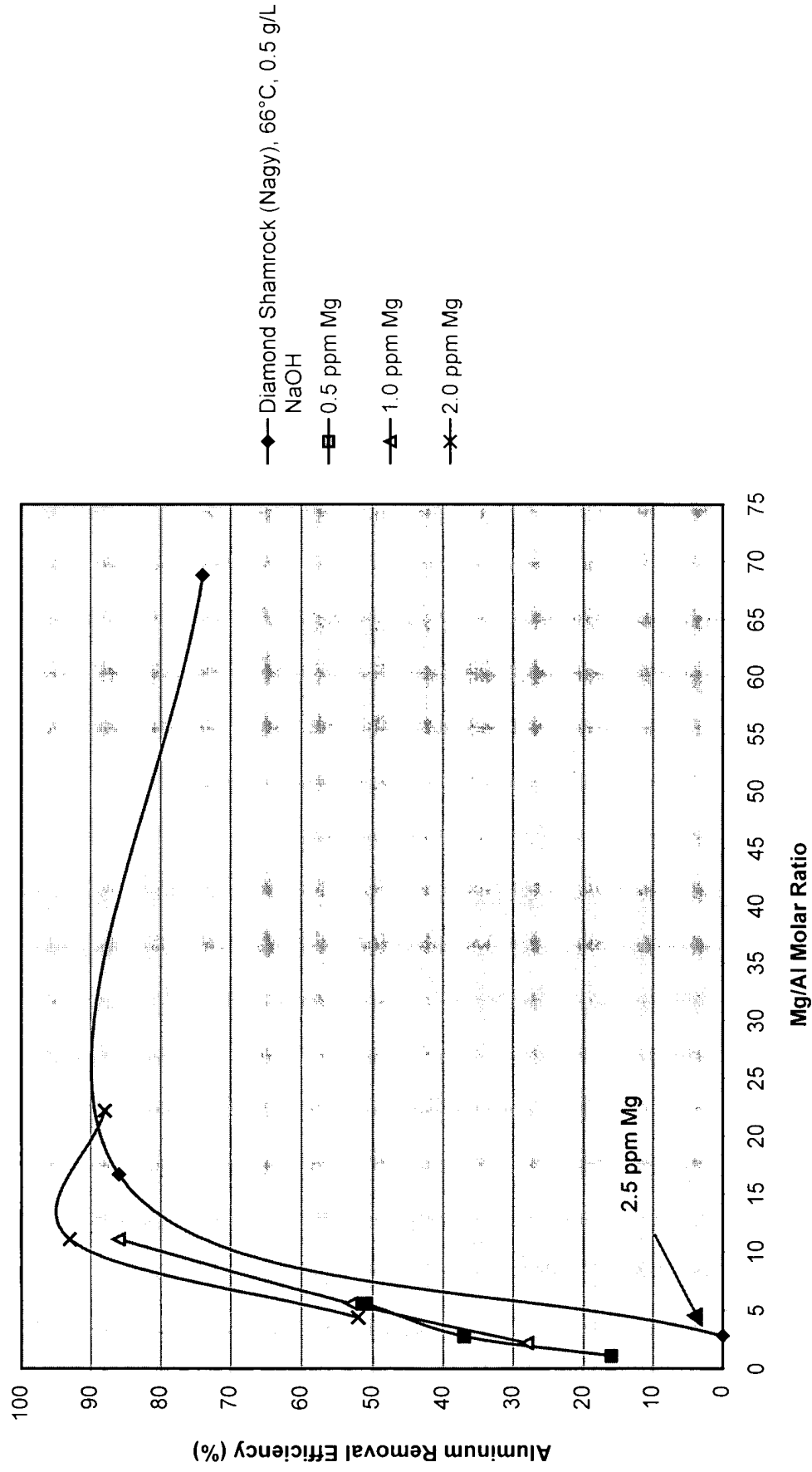
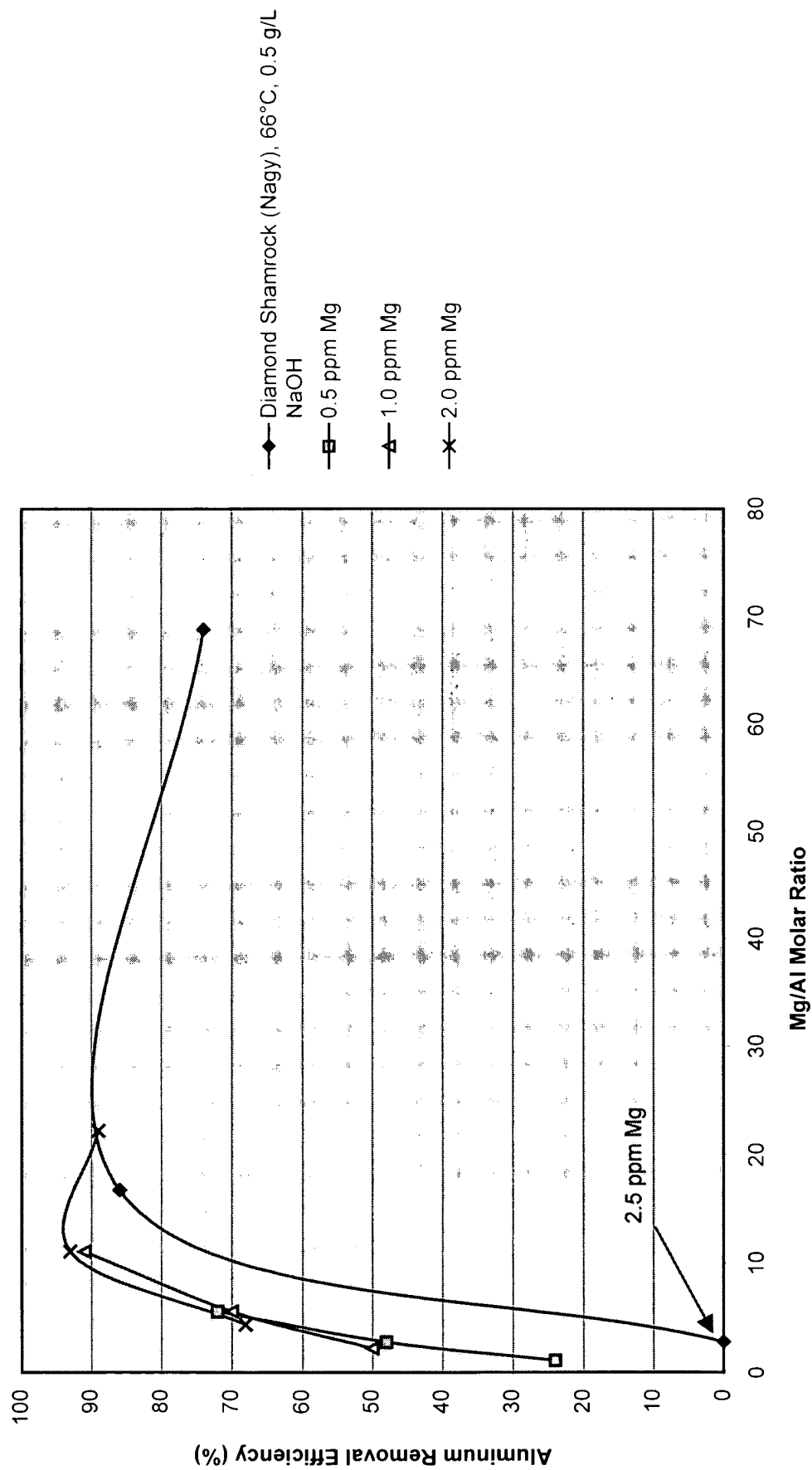


Chart 2 - Comparison of Aluminum Removal Efficiency (%) at Various Mg/Al Ratio with Low Magnesium Concentration
 Temperature 50°C; Excess Alkalinity Level at 0.1 g/L NaOH



Date: F b 13/02

The following results have been tabulated to illustrate the effects of Aluminum removal using Magnesium at low concentration.

Temperature: 50-60°C Brine: Ion-Exchange Quality Excess NaOH : 0.1-0.2 g/L
Residence Time: 5-15 minutes

Experiment #1

Initial Aluminum Concentration (ppm)	Magnesium Concentration Added (ppm)	Mg/Al Molar Ratio	Residual Aluminum (ppm)	Al Removal Efficiency (%)
0.5	3	6.7	0.010	98
0.5	4	8.9	0.001	99
0.5	5	11.1	0.001	99

Experiment #2

Initial Aluminum Concentration (ppm)	Magnesium Concentration Added (ppm)	Mg/Al Molar Ratio	Residual Aluminum (ppm)	Al Removal Efficiency (%)
1	1	1.1	0.840	16
1	2	2.2	0.650	35
1	3	3.3	0.430	57
1	4	4.4	0.210	79
1	5	5.5	0.010	99